SELENIUM-79 IN SOILS AND REACTOR WASTE STREAMS

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ABSTRACT:

Selenium-79 is a long-lived man-made radionuclide which is produced as part of the nuclear fuel cycle. It is a low abundance fission product with a half life of 65,000 years and decays by weak beta emission with no gammas or x-rays. Capability to analyze samples for selenium is often required as a component of remedial investigations, environmental assessments, remediation contracts, or reactor waste stream characterization.

The Thermo NUtech laboratory in Richmond, CA has developed a procedure for analyzing Se-79. This procedure has proven applicable to both environmental samples as well as highly radioactive reactor waste stream samples. Chemical yields, based on weighing added stable selenium carrier, range from 70 to 80% on soil and reactor waste stream samples. This paper presents the salient features of sample digestion/dissolution, purification of the Se fraction and the creation of the final Se metal fraction which is dissolved for measurement by liquid scintillation counting.

PROCEDURE:

1. Dissolution or Digestion - The aliquot is transferred quantitatively to a 250 mL distillation flask. A standardized Se carrier along with holdback carriers such as Co, Fe, and Cs are added. To the mixture is added 15 mL of 1:1 ratio of HNO_3 - $HCLO_4$, 1 mL of 6N HCl, and 3 mL of DI water. The mixture is digested at 70 - 90°C until the heavy white fumes of perchloric acid are evolved. HNO_3 presence is tested to assure that no trace of HNO_3 is left in the solution prior to the distillation step.

2. Distillation - 10 mL of HCl and 5 mL of 48% HBr are added into the distillation flask. Distillation temperature is raised gradually and helium is used as the carrier gas. The $SeBr_3Cl$ distillate is collected in a receiver submerged in an ice bath.

3. Precipitation - the pH of the distillate is adjusted to 1.0 with HCl or with NH_4OH. $NH_2OH.HCl$ is added to reduce the selenium complex into Se metal with heating. The metal is separated by centrifugation. The precipitate is dissolved in 0.5 mL of HNO_3 and a scintillation cocktail is added. The final cocktail is sent to the radiation measurements laboratory for beta counting.

FACTS AND FIGURES

From October 1993 through November 1994, 127 soil samples and 47 water samples were processed using this procedure.

During the first six months of using the procedure we were getting a yield higher than 90%. We were also experiencing a quenching problem and this was traced to tramp salts precipitating during the reduction of the selenium/bromochloride to selenium/metal due to prolonged heating. We soon realized that a yield higher than 90% creates an artificial recovery that results in quenching of the selenium carrier.

In March 1996 the reduction process was change to keep the maximum heating time to one hour at 70 - 90°C, or to half the volume of the solution. This solves the quenching problem and the recovery then ranged from 40 to 85%.

In general, including the chemical yields that were below 30% and over 90% the average recovery is 64% ±17%. Excluding yields below 30% and those of over 90% gives us an average yield of 65% ±14%.